1. Introduction

Femtosecond laser filamentation is a unique nonlinear optical phenomenon during which an intense laser pulse could self-focus and propagate inside a transparent optical material without significantly losing its high intensity over a long distance [1–8]. At the end of the filament, the pulse self-transforms into a chirped white light laser pulse [7].

T Zeng1, J Y Zhao1, W Liu1 and S L Chin2

1 Institute of Modern Optics, Nankai University, Key Laboratory of Optical Information Science and Technology, Ministry of Education, Tianjin 300071, People’s Republic of China
2 Département de physique, de génie physique et d’optique, Centre d’optique, Photonique et Laser (COPL), Université Laval, Québec, QC G1V 0A6, Canada

E-mail: liuweiwei@nankai.edu.cn

Received 16 February 2014, revised 25 April 2014
Accepted for publication 27 April 2014
Published 28 May 2014

Abstract

Angular distribution of the backscattered nitrogen fluorescence induced by femtosecond laser filamentation in air has been studied. The experimental results demonstrate that the fluorescence intensity forms multiple rings and is amplified in the backward direction through amplified spontaneous emission (ASE). More importantly, the ASE signal features an angular dependent gain coefficient. The results are valuable for the optimization of the remote-sensing setup by the filamentation in air.

Keywords: ultrafast, filamentation, remote sensing

(Some figures may appear in colour only in the online journal)
was realized by focusing an ultraviolet (266 nm) laser pulse in air, whose duration is 100 ps. The lasing occurs at 845 nm and the estimated divergence is about 40 mrad, which is consistent with diffraction-limited lasing from the cross-sectional size of the pump volume.

In the present work, the transverse intensity distribution of the backward propagating air lasing at 357 nm has been measured. The air laser was induced by the femtosecond laser filamentation process. The experimental results indicated that in the backward direction, the spatial profile of 357 nm light consists of multiple rings. The recorded divergence exceeds 15°. At the same time, the effective backward gain has strong spatial dependence. The inner ring has higher gain than the outer ring.

2. Experimental setup

The experimental setup is sketched in figure 1. In brief, a 1 kHz, 800 nm, 50 fs (FWHM) Ti:sapphire laser pulse was focused by an \( f = 100 \text{ cm} \) lens in air. The initial beam diameter is about 0.8 cm (1/\( e^2 \)). The filament was generated near the geometrical focus. The backscattered fluorescence from the filament in air was then detected by using a photomultiplier tube (PMT), which was set behind the last 1 inch diameter sending mirror M1. Note that the last sending mirror was stuck on the top edge of a mirror mount in order to avoid the clipping of the signal by the mirror mount. Furthermore, an iris with 5 mm diameter aperture was inserted in front of the PMT to limit the field of view. The fluorescence signal was then recorded with a fast oscilloscope. A dielectric coated fused silica mirror (reflectivity: >99.9% at 0° incident angle, central wavelength: 800 nm; bandwidth: around 100 nm) was put before the PMT in order to filter out the scattered fundamental laser light. The detected light wavelength was selected by an appropriate interference filter with a bandwidth of 3 nm at the central wavelength of 357 nm, which corresponds to the (1–0) vibronic transition of the second positive band system of N₂ [28].

To measure the angular distribution of the fluorescence signal, the PMT was put on a translation stage that can move in the direction perpendicular to the laser propagation direction. When the PMT was set at the backward direction (0°), the distance between the PMT and the geometrical focus was about 110 cm. The maximum displacement of the translation stage was 54 cm. By taking the geometrical focus as the reference point, the scan range corresponded to ±15° in angle.

3. Results and discussion

The filament started before the geometrical focus and the filament length increased towards the focusing lens with increasing laser energy. During the experiment, we took the integral of the whole area under the signal peak from the PMT as the fluorescence intensity. The background DC dark current of the PMT has been subtracted from the signal. The PMT was moved along the translation stage with a step of 5 mm (around 0.26–0.28° in angle depending on the PMT position) to measure the 357 nm light signals from different angles. For each angle, the variation of the fluorescence signal intensity \( S \) was recorded with respect to the pumping laser energy. The data acquisition program registered the laser energy and the induced fluorescence signal of the same shot simultaneously. After that, the signals at different angles were analyzed for the same input energy. The outcome is shown in figure 2 for three representative pumping laser energies: 5, 10, and 15 mJ, respectively. Note that since the signal around 0° passed through the last sending mirror before it reached PMT, the data points within ±0.5° (corresponding to the mirror dimension) have been corrected by the transmission curve of the last sending mirror.

In figure 2, the angular dependence curves for different input laser energies are similar. The curves are rather symmetrical centred at 0°. The small asymmetry could be due to the nonperfect alignment of the translation stage. It is also clearly shown that most of the fluorescence signal concentrates in a region of around ±10° from the central axis. Moreover, there are some fine peaks shown in the plot. These peaks correspond to the ring structure in the spatial distribution if we transfer it to three dimensions. The maximum signal was found when the displacement of the PMT was...
4.8 cm. The corresponding divergent angle is thus given by: \[ \theta = \arctan\left(\frac{5.0}{110}\right) \approx 2.5^\circ (~46 \text{ mrad}) \]. This value is in good agreement with the previously estimated divergent angle of the forwardly emitted air laser [20, 24, 27]. However, the signal decreased 1/3 just around the 0° with respect to the maximum signal recorded at ±2.5°. Furthermore, in figure 2, the fluorescence intensity gets down to a minimum around ±9.5° (\( \theta = \arctan(18.5/110) \)). The signal increases again and reaches the second maximum at ±11° although the signal at this angle is five times weaker than the first maximum. Then it decreases to the minimum at about ±15°, which is the limit of the detection system. The angle corresponding to the second maximum for lower laser input energy is larger than that for higher energy.

We further look into the laser energy dependence of the fluorescence signal for certain angles. Figure 3(a) shows the evolution of the fluorescence signal as a function of input laser energy for detection angles of 0, 2.5, and 11°, respectively. The three curves all increase significantly with higher laser energy. Since the emission of the nitrogen fluorescence originates from the excitation and ionization processes of N₂ molecules [29], the trend of signal \( S \) depicted in figure 3(a) could be potentially induced by the variation of the laser intensity inside the filament. Previous work has suggested that the probability of multiphoton ionization to N₂⁺ approximately goes with the 8th power of the laser intensity [30], i.e., \( S \propto I^8 \) or \( S \propto I^8 \). For the sake of verifying the spanning range of the laser intensity required to induce the fluorescence change shown in figure 3(a), we illustrate in figure 3(b) the variation of \( \sqrt[8]{S} \) versus the input laser energy. As indicated in figure 3(a), \( S_{\text{tot}}/S_{\text{tot}} \approx 260 \), yielding \( I_{\text{sat}}/I_{\text{sat}} \approx 2 \) as shown in figure 3(b). However, it has been demonstrated that due to intensity clamping, the laser intensity could not be enhanced dramatically [31–35]. Therefore, in order to explain the experimental results of figure 3(a), we will consider that the fluorescence signal increase is mainly caused by the lasing effect inside the filament [12]. It is worth mentioning that Jin et al. have measured the angular distribution of the backward supercontinuum emission from a filament in air [36]. They have shown that the supercontinuum is peaked around 0° and confined within a cone angle of 10° in the backward direction.

**Figure 3.** (a) The fluorescence intensity of N₂ at 357 nm (S) as a function of laser energy at different angles. (b) \( \sqrt[8]{S} \) as a function of laser energy at different angles.

Thus, as suggested by [12] the fluorescence intensities versus filament length at several different angles are shown in figure 4. The filament length is converted by taking the difference between the self-focusing position of peak power and the geometrical focusing of the external lens [12]. The plot for the positive angle has been shifted vertically in order to avoid the overlap. The data do not extend to the zero filament length because the fluorescence signals are too weak to be detected when the input energy is lower than a certain value. All the
curves show a similar tendency even though the signal intensity differs for different angles. And the curves for the symmetrical angles are rather similar.

According to Luo et al [12], the effective backward ASE gain could be described by:

\[ I \propto P = \int_{0}^{L} P e^{\theta} d\theta = \frac{P}{g} (e^{\theta} - 1) \]  

(1)

where \( P \) denotes the spontaneous emission power per unit length, \( L \) corresponds to the estimated length of the filament, and \( g \) is the effective gain coefficient. Considering that the intensity is clamped down inside the filament [31–35], we assume the spontaneous emission power \( P_s \) is uniform for every angle. In figure 4, the fitted curves are plotted as solid lines by using \( P_s \) and \( g \) as fitting parameters.

The fitted gain coefficient is different for different angles. In figures 4(a) and (b), the gain coefficients are close (around 0.22 cm\(^{-1}\)) but changed a bit from 0 to ±2.5°. The fluorescence propagating along 0° has been less amplified compared with those along the ±2.5° direction. Compared with figure 4(c), the gain values for these three angles are much larger than that for the outer region at ±11° (around 0.13 cm\(^{-1}\)). As shown in figures 4(b) and (c), the gain coefficients are also symmetrical for the symmetrical angle.

Several previous works [37–42] have shown that a complicated ring structure could be formed for the fundamental laser beam pattern during the propagation. The transverse fluence profile showed that there was a dip in the center surrounded by several outer rings. In our experiment, beam profiles inside the filament have also been investigated. Two parallel fused silica wedges were inserted in the laser beam path, both at grazing angles, yielding a reflectivity of about 10% at each front surface. Therefore, after two surface reflections, the laser intensity was reduced to approximately 1%. The cross sections of the laser beam were then registered by a CCD camera. Various neutral density filters were put in front of the CCD camera to further attenuate the laser intensity. In order to avoid the damage of the wedges, the beam profiles were only recorded at low energy, namely, 1.5 mJ. Figure 5 demonstrates

![Figure 5. Representative beam profiles recorded at three distances inside the filament: (a) \( z = -2 \) cm, (b) \( z = 0 \) cm, and (c) \( z = 2.5 \) cm. The input energy is 1.5 mJ.](image-url)
the fluorescence signal would weakly depend on the focal length and initial laser beam diameter in the application of the filamentation-based remote sensing.

4. Summary

In conclusion, the angular distribution of the scattered fluorescence shows significant enhancement in the backward direction. The fluorescence goes through a different amplification due to the ring structure of the intensity distribution inside the filament. The knowledge about the backward angular distribution of air lasing could be very important for the remote-sensing technique, particularly for optimizing the detection geometry of LIDAR setup, such as the view angle and the size of the collecting optics.

Acknowledgments

This work is financially supported by National Basic Research Program of China (2014CB339802, 2011CB808100) and National Natural Science Foundation of China (11174156). W L acknowledges the support of the open research funds of State Key Laboratory of High Field Laser Physics, Shanghai Institute of Optics and Fine Mechanics (SIOM). The financial support of the Natural Sciences and Engineering Research Council of Canada and of the Canada Foundation for Innovation are also acknowledged.

References

[27] Dogariu A, Michael J B, Scully M O, Miles R B 2011 Science 331 442